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Delivering science and technology to protect our nation and promote world stability

Nuclear Forensics at Los Alamos National Laboratory

Los Alamos, NM

William Kinman, Robert Steiner, Stephen Lamont

October 19, 2016

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NNSA
National Nuclear Security Administration

Operated by Los Alamos National Security, LLC for the U.S. Department of Energy's NNSA

Outline

- **Nuclear Forensic Science Overview**
- **Investigations and Expertise**
- **Common Analysis Techniques**
- **National Nuclear Forensics Library**
- **Case Study:** *Bulgarian HEU seizure*
- **Case Study:** *Origins of Pu in the Environment*
- **Case Study:** *Post detonation Nuclear Forensics*
- **Summary**

Nuclear Forensic Science

Nuclear forensics is the collection and analysis of nuclear or radiological material to support investigations into the diversion, trafficking, or illicit activities involving such materials.

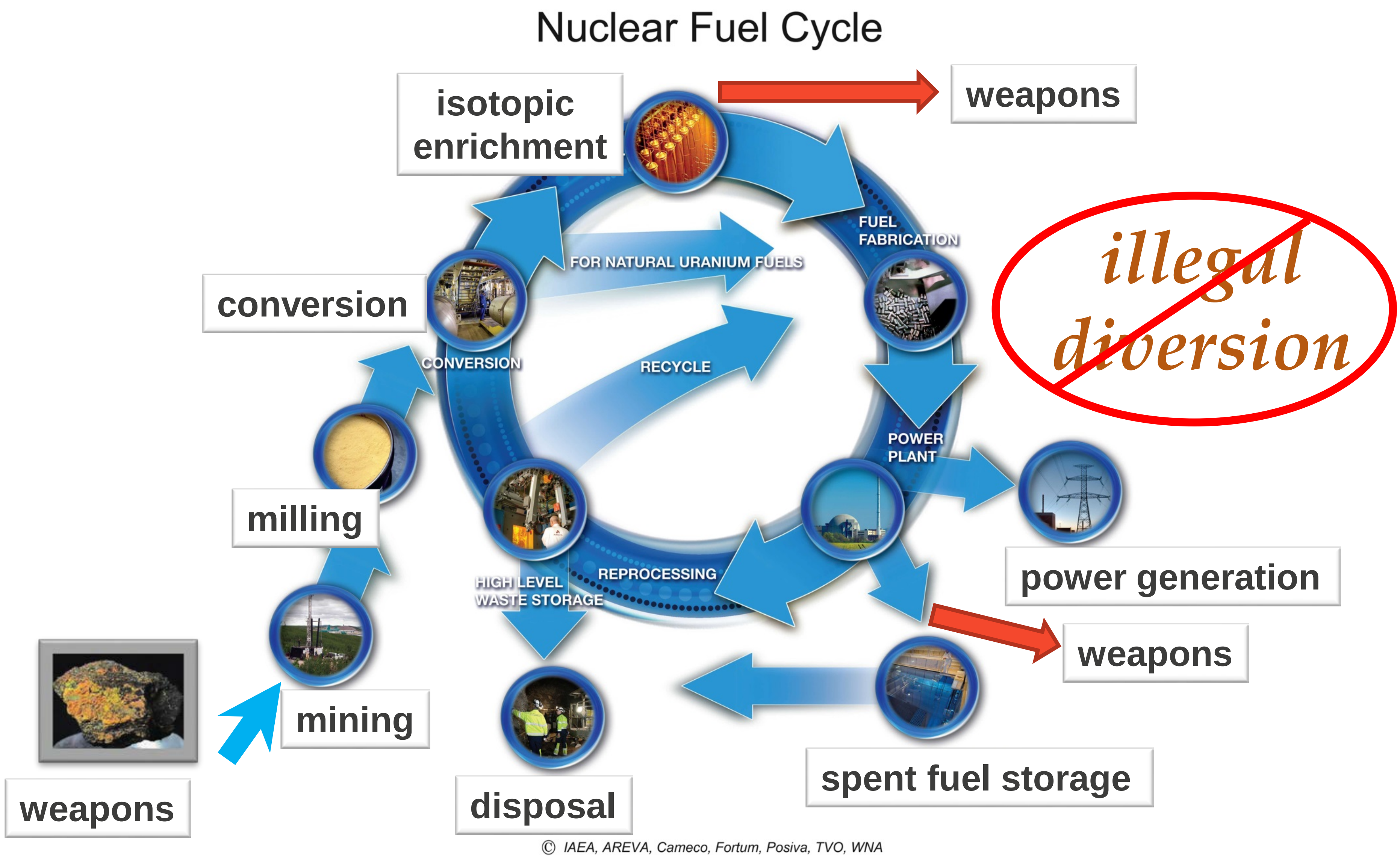
Goal: Link nuclear or radioactive materials to people, processes, events and/or locations

Nuclear Power ***Pre-Detonation***

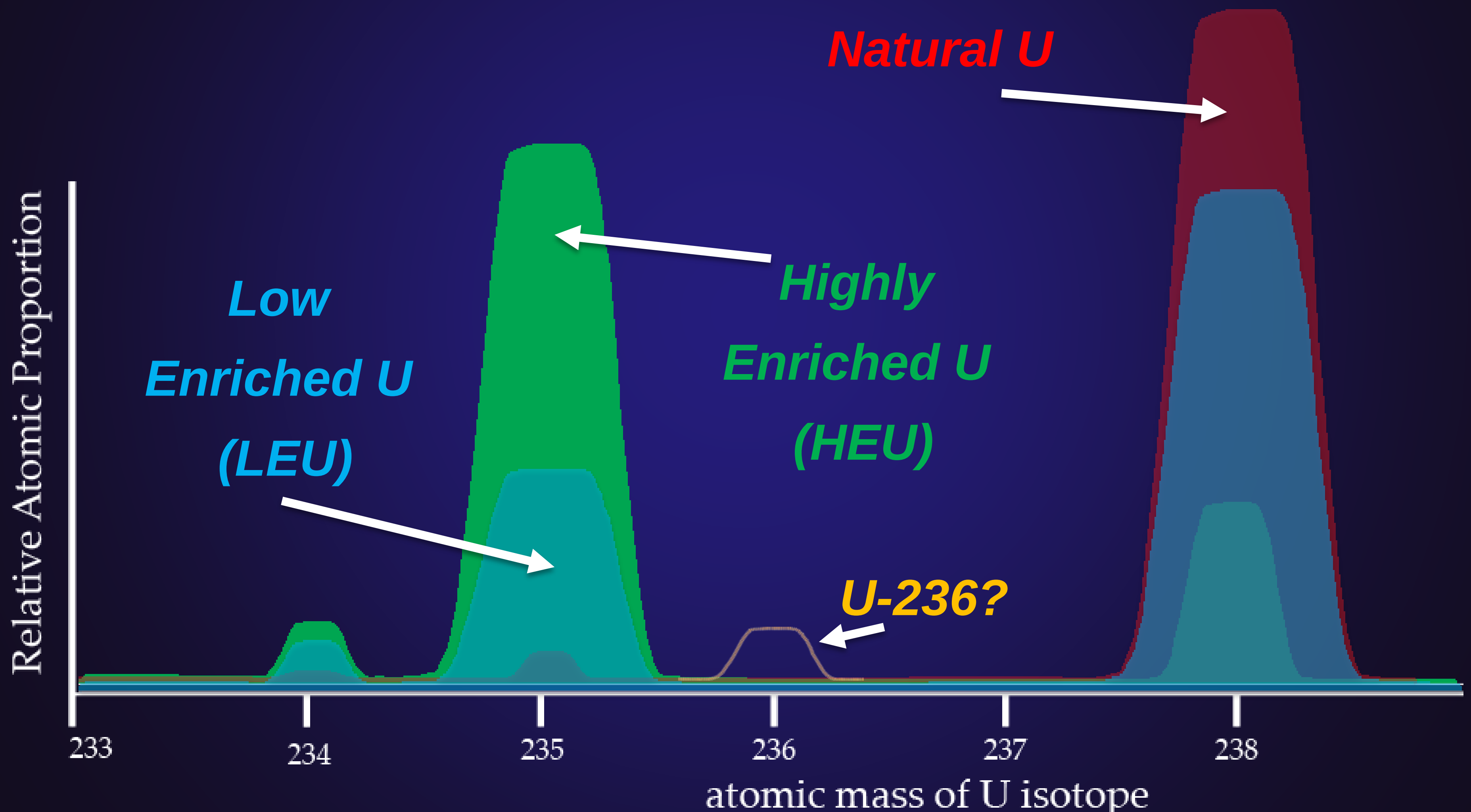
Nuclear  ***Materials***

Radiation Sources ***Post-Detonation***

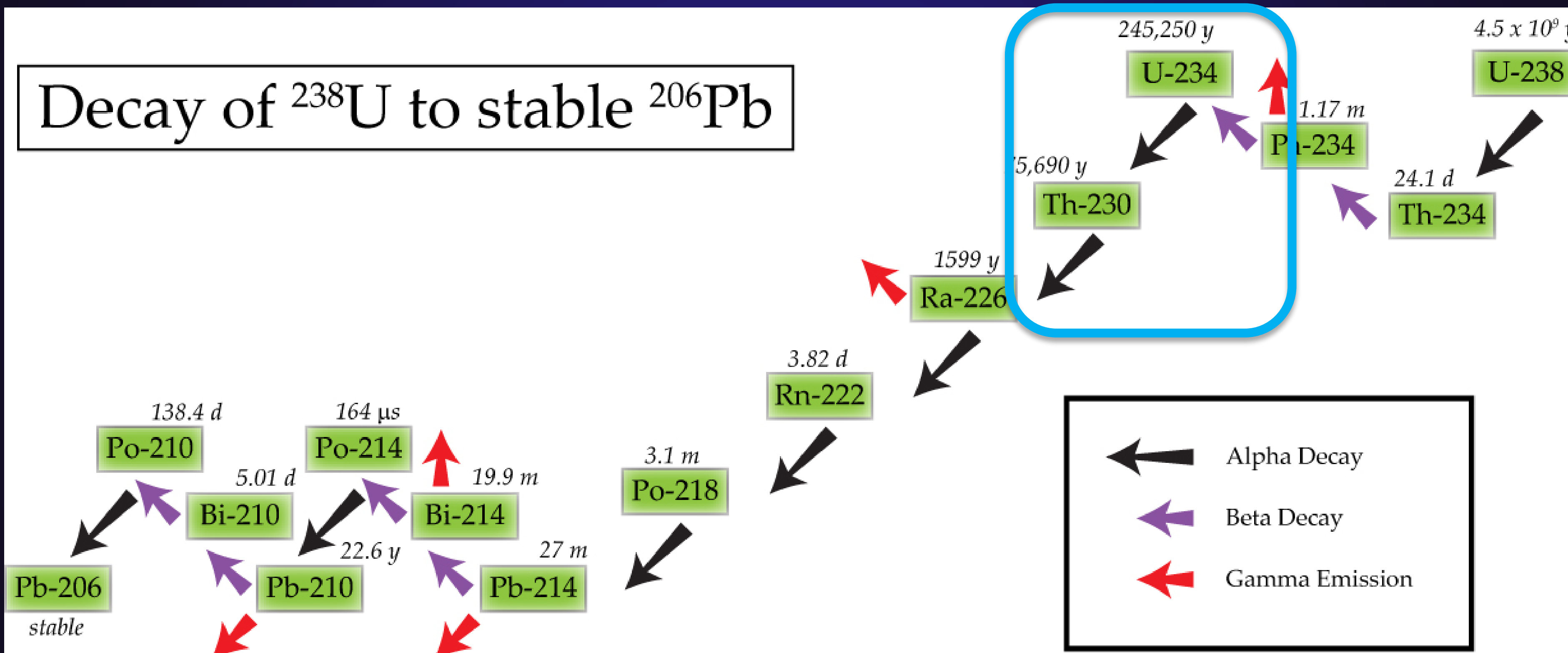
Nuclear Fuel Cycle



Uranium Isotopic Composition Primer

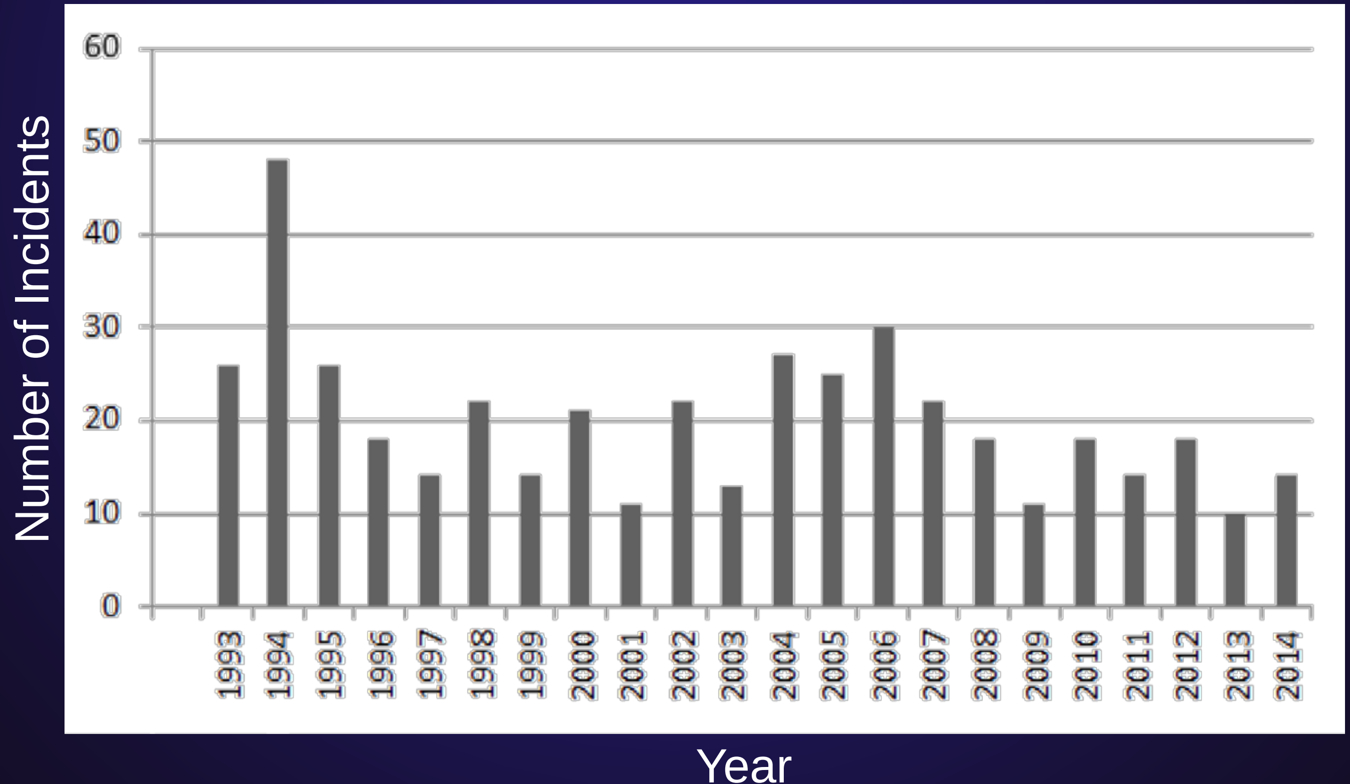


Radiochronometry Primer



The Importance of Nuclear Forensics

International Atomic Energy Agency's (IAEA's) Confirmed Cases of Unauthorized Possession and Criminal Activity involving Nuclear and Radioactive Materials

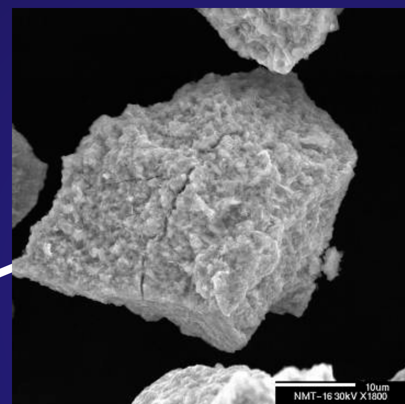


The Importance of Nuclear Forensics

- **Nuclear forensic techniques are also used to support regulatory investigations involving nuclear and radioactive materials**
  - Origin of radionuclide contamination in the environment
  - Provenance of orphaned radioactive sources
  - Sources of radionuclides involved in internal exposure claims

Nuclear Forensics Objectives

To perform forensic analysis on nuclear materials by identifying key elements for forensic investigations



Unknown Nuclear Material

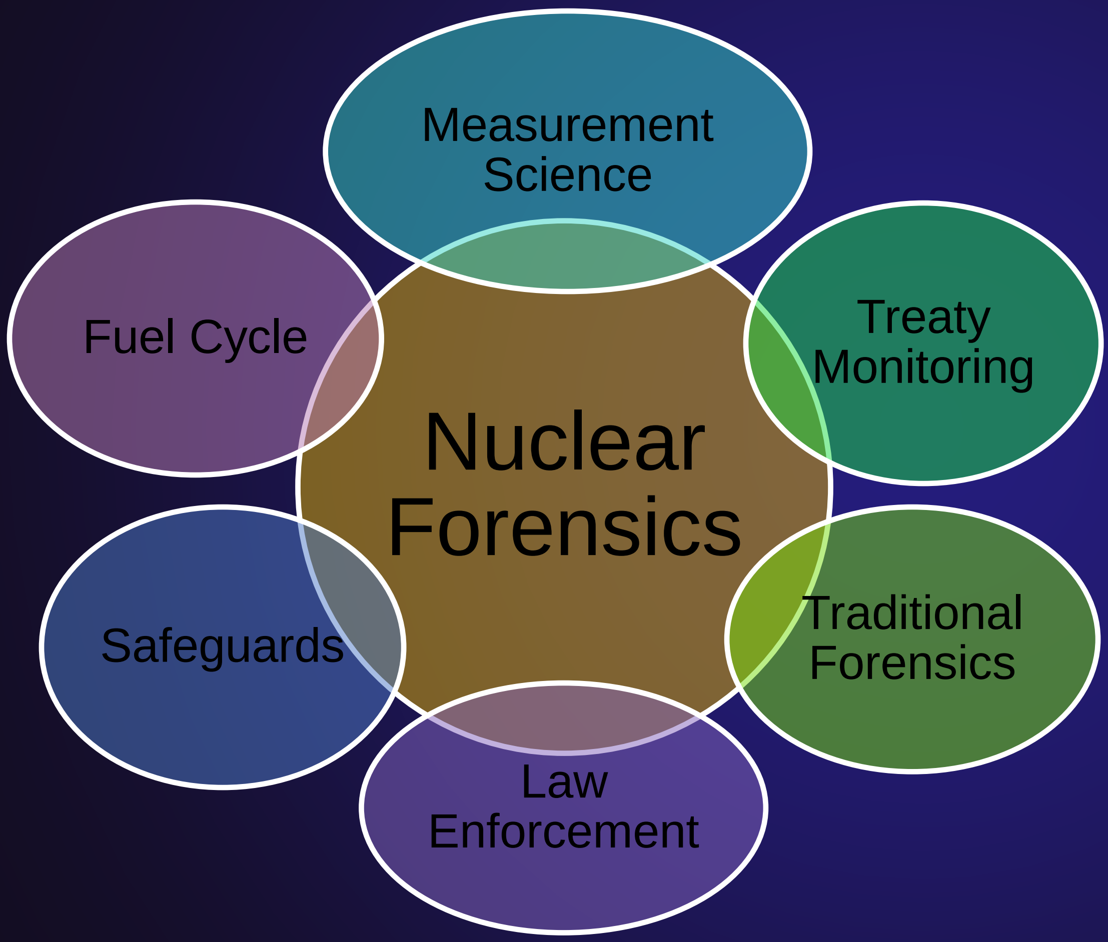
How was it produced?

Where was it produced?

Date of Production?

What was its intended use?

Nuclear Forensics Expertise



Many Disciplines Contribute

- Radiochemists
- Geochemists
- Analytical chemists
- Reactor physicists
- Nuclear engineers
- Process engineers
- Enrichment engineers
- Statisticians
- Quality assurance

US DOE National Laboratories

Our ability to characterize nuclear materials and processes relies on analytical methods from:

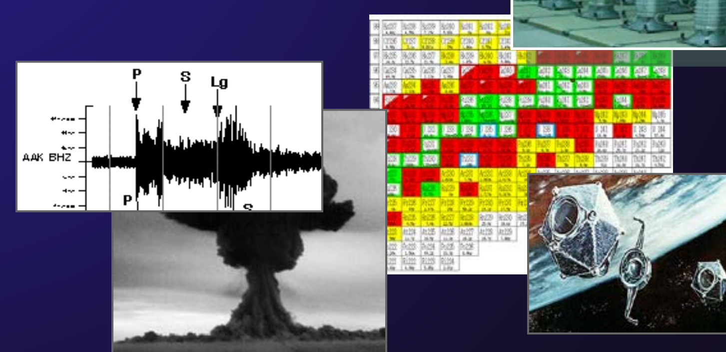
Certification of materials in pit production

Our ability to characterize signatures of worldwide nuclear materials production relies on skills built in:

Support for international safeguards programs

Our ability to characterize the origins of a nuclear explosion is based on:

Underground test experience



LANL facilities to work with materials of all quantities: *example Pu-239*

Low-Level ⟷ High-Level

< 10^{11} atoms Pu-239

TA-48

10^{9} – 10^{22} atoms Pu-239

Nuclear facilities (CMR, PF-4, RLUOB)

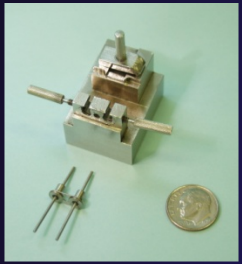
> 10^{22} atoms Pu-239

TA-55

All facilities house ongoing missions that exercise analytical capabilities routinely

High-level Actinide Analysis Capabilities

Mass Spectrometry

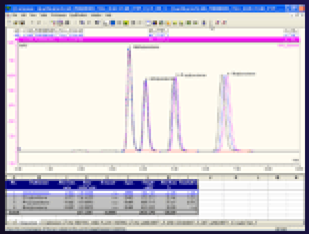


High-Precision
Thermal Ionization Mass Spectrometry

Radiochemistry and Nondestructive Analysis

Alpha and gamma spectrometry
Gross alpha, liquid scintillation

Interstitial Analysis & Ion Chromatography



Fluoride, chloride, nitrite
nitrate, phosphate, sulfate
oxalate and perchlorate

carbon, oxygen, hydrogen
sulfur, moisture, and tritium

High-level Actinide Analysis Capabilities

Onsite Analytical Chemistry and Sample Management

Coordinate sample receiving, shipping, and distribution at TA-55 and CMR

Onsite radiochemical and trace analysis

Assay and Classical Chemistry

Coulometric titration
Ceric titration
Pu (III) and Pu (IV)
U Assay by Davies Gray
Fe and Si determination
Loss on Ignition (LOI)
Free acid determination
Standard preparation

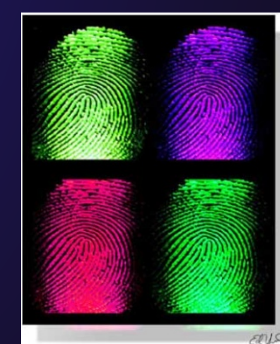
Plasma Spectroscopy

Inductively Coupled Plasma-Mass Spectrometry
Inductively Coupled Plasma- Atomic Emission Spectrometry

DC Arc Emission

Cold-Vapor Atomic Fluorescence

X-Ray Fluorescence (XRF) and X-Ray Diffraction (XRD)



Fingerprint Detection Technology

Low-level Non-Destructive Analysis Tools

The 7000 ft^2 counting facility located at TA-48 at LANL is equipped with 80+ gamma spectrometers, 100+ alpha spectrometers, 2 liquid scintillation counters, 6 automatic beta counters, a high purity Ge Clover detector, and digital autoradiography. The TA-48 counting facility makes more than 50,000 measurements each year.

The high purity Ge Clover Detector is a high efficiency, low background detector system with active background suppression and event-by-event data capture

Low-Level Destructive Analysis Tools

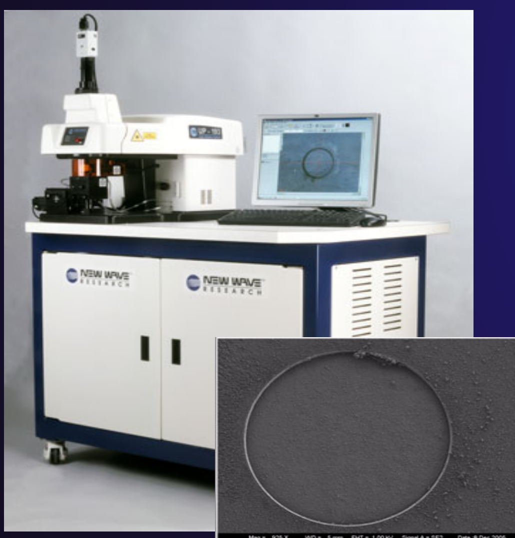
Multi-collector ICP-MS
(MC-ICP-MS)
High precision, high accuracy
Isotope ratios (U, Sr, Pb, Fe, B…)
ng to <fg sample requirements

Sector Field ICP-MS
(SF-ICP-MS)
Ppq – ppm elemental
concentrations

Multi-collector
Thermal Ionization MS
(TIMS)
Pu, other actinide, Sr, Nd

In-Situ Analysis Tools

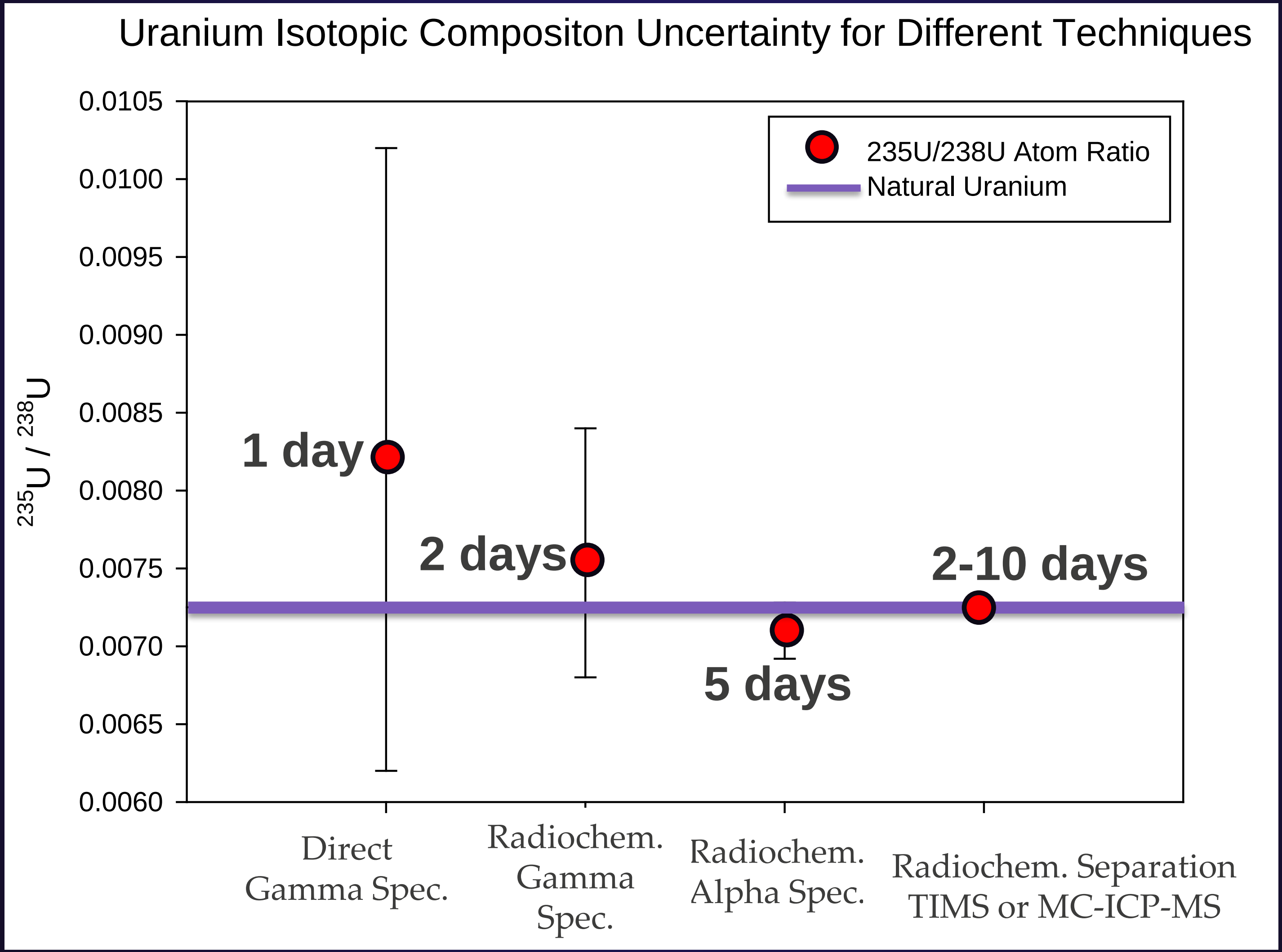
Laser ablation
193 nm ArF Excimer
In-situ analysis w/
ICP-MS systems
Few micron spatial
resolution



Field Emission
Environmental SEM
(FE-ESEM))
Morphology
Major, minor elemental
characterization w/
WDS, EDS systems

Cameca 1280
High transmission,
High sensitivity
Secondary
Ionization MS
(SIMS)

Precision and Timeline for Nondestructive and Destructive Analysis Methods



Nuclear Forensics: Evidence

Part 1: Traditional Forensics:
Link individuals to criminal activity

Material Analysis

Judicial Proceedings

Trafficker Convicted

- Led by federal law enforcement
- Important for criminal proceedings
- Requires high-quality, legally defensible analyses
  - What is it?
  - How much is there?
  - Was a law violated?
- Does not require a detailed analysis of all material attributes
- ***Most countries have the technology, equipment and expertise for these analyses***

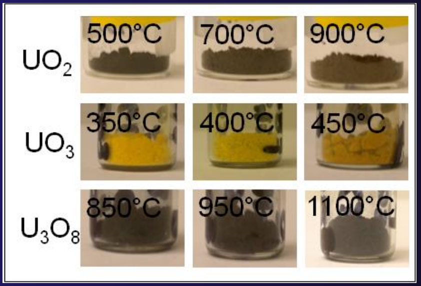
Nuclear Forensics: Investigations

- Detailed analysis of materials –
  - Los Alamos National Laboratory
  - Lawrence Livermore National Laboratory
- Expert evaluation and comparative analysis
- Assessment of material origin
- ***Requires advanced capabilities:***
  - ***Laboratory analysis***
  - ***Data interpretation***
  - ***National Nuclear Forensics Library***

Part 2: Investigative Forensics:
History of nuclear material

Full Characterization
- Precision isotopics
- Chemical composition
- Age dating
- Morphology

Comparative Analysis
- Intended use
- Process history
- Fuel cycle information



Outcome
- Possible origins
- Connections between cases
- Enhanced security

Important Investigative Question: Is it ours?

- Nuclear material found outside of administrative control: “Is this consistent with our material?”

- States have a responsibility to identify materials found out of regulatory control and determine if they are consistent with those used, produced, or stored within their borders

- A National Nuclear Forensic Library is extremely valuable for answering this question with timeliness and confidence

National Nuclear Forensics Library Model

- A **National Nuclear Forensics Library** is a national system of ***expertise*** and ***information*** necessary to identify nuclear or other radioactive material found out of regulatory control

National Nuclear Forensics Library	≠	Database
National Nuclear Forensics Library	=	Forensics Experts + Material Experts + Data Resources

NNFL Effort and Complexity

- **NNFL complexity is largely dictated by the nuclear activities within a state**

Complexity of NNFL and Associated Materials Databases $\propto$

- Quantity and variety of radioactive sources
- Quantity and variety of nuclear materials
- Production or processing of nuclear or radioactive materials
- Research and development activities

Not every country needs to capture the same material characteristics to have a functional NNFL

Case Study #1

Highly Enriched Uranium seized in Rousse, Bulgaria on May 29, 1999 at Romania-Bulgaria border crossing

** see more: Niemeyer, S., & Hutcheon, I. (2002). Forensic analysis of a smuggled HEU sample interdicted in Bulgaria (IAEA-CN--98). International Atomic Energy Agency (IAEA)

1999 Bulgaria 73% HEU Example**

Non-nuclear forensics

- Wax type
- Wax colorant
- Paper origin
- Pb metallurgy
- Pb isotopics
- Ampoule material

Nuclear material forensics

- Particle characterization
- Stoichiometry
- Impurity elements
- Residual radionuclides
- Age-dating
- U & Pu isotopics

LLNL-Led Effort: Excellent demonstration of what can be done!

** see more: Niemeyer, S., & Hutcheon, I. (2002). Forensic analysis of a smuggled HEU sample interdicted in Bulgaria (IAEA-CN--98). International Atomic Energy Agency (IAEA)

1999 Bulgaria 73% HEU Summary**

- Primarily U_3O_8; 72.7% U-235, 12.1% U-236
- Reprocessed irradiated material
- 3 ppb Pu ($^{240}Pu/^{239}Pu = 0.12$)
- Impurity and radiochronometry results indicate that batch Purex reprocessing
- Mean date of chemical reprocessing = October 30, 1993 +/- 25 days

** see more: Niemeyer, S., & Hutcheon, I. (2002). Forensic analysis of a smuggled HEU sample interdicted in Bulgaria (IAEA-CN--98). International Atomic Energy Agency (IAEA)

Case Study #2

Plutonium identified in soil samples from a mountain range in northern New Mexico USA

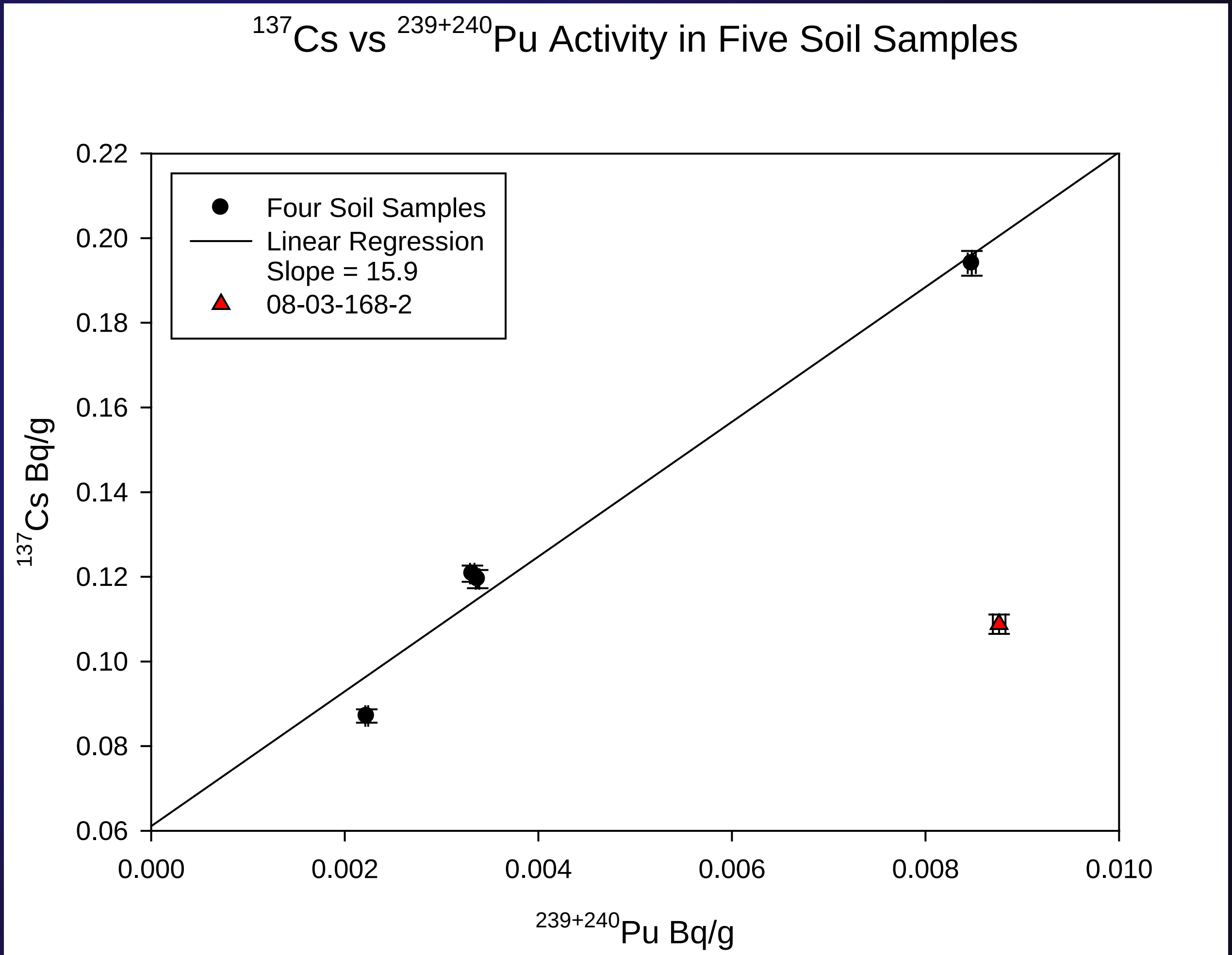
Investigating Origins of Pu in the Environment

- **Claim:** Plutonium from LANL was contaminating the Sangre de Cristo mountains, which are downwind from LANL
- **Question:** Is the Pu in this environment consistent with LANL Pu or other sources?

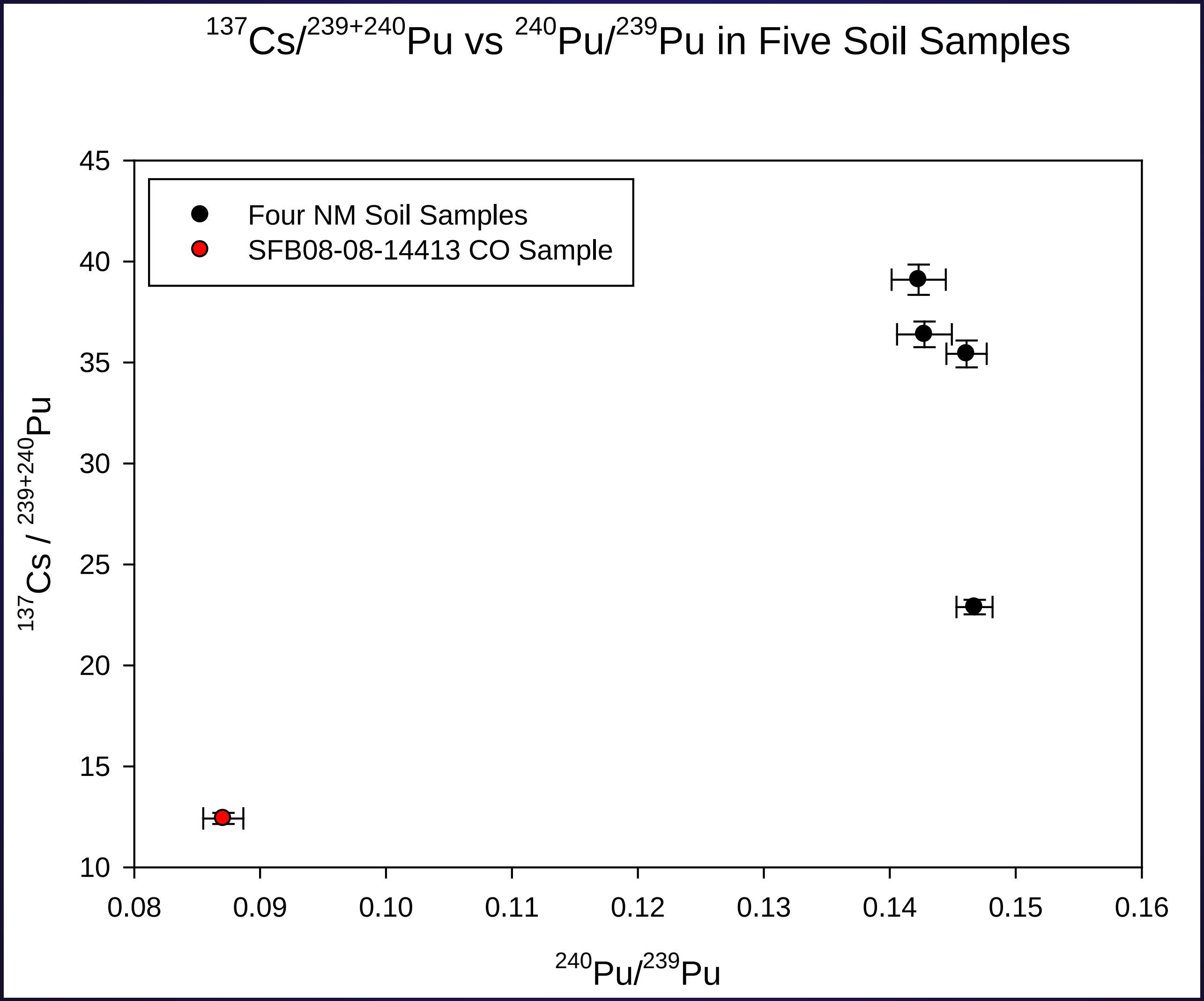
Results: ^{137}Cs and $^{239+240}$Pu Activities

Provenance of Pu assessed in 5 soil samples based on:

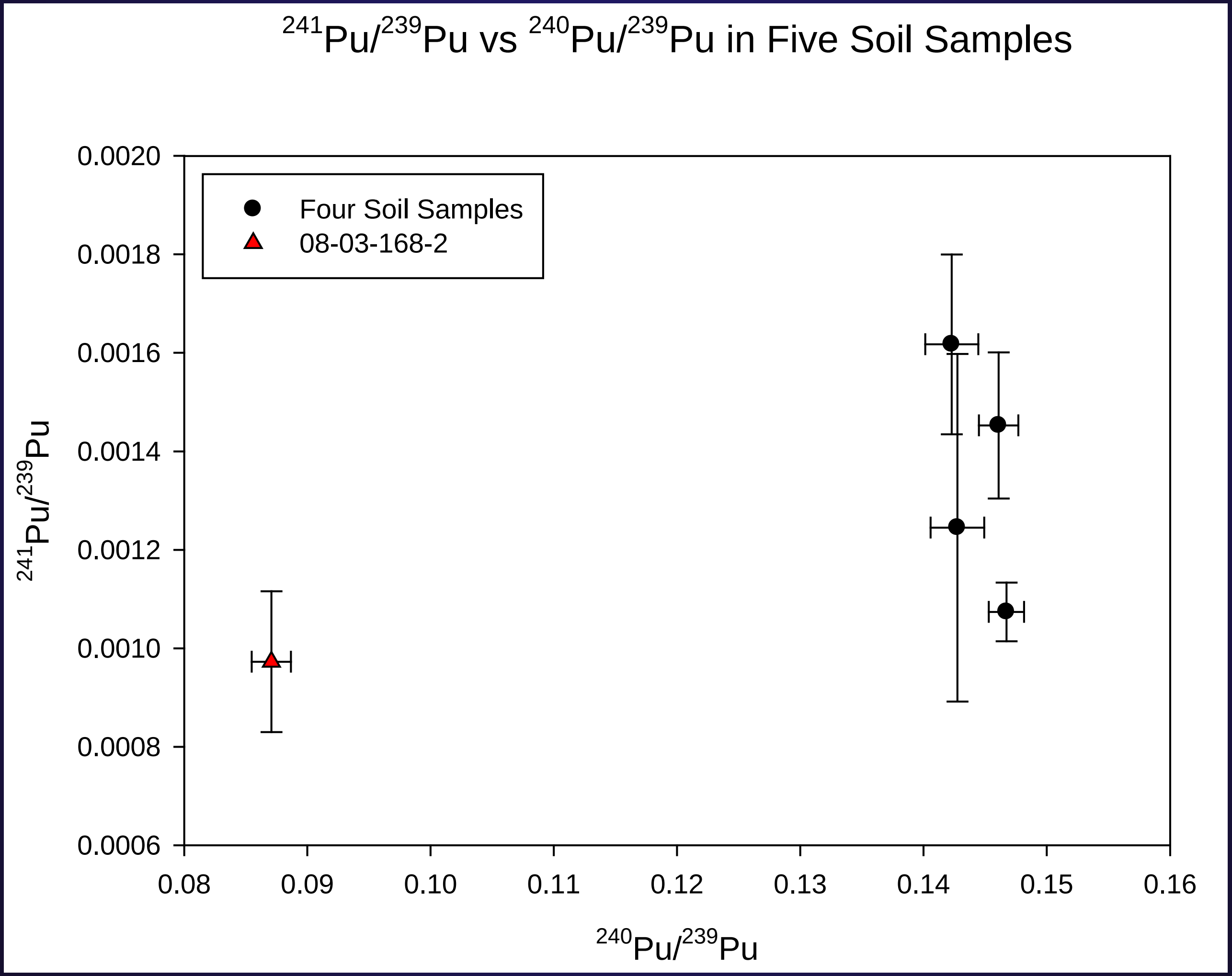
- ^{137}Cs activity
- $^{239+240}$Pu activity
- ^{240}Pu/^{239}Pu
- ^{241}Pu/^{239}Pu



Results: ^{137}Cs / $^{239+240}Pu$ vs. $^{240}Pu/^{239}Pu$



Results: Pu Isotope Ratios



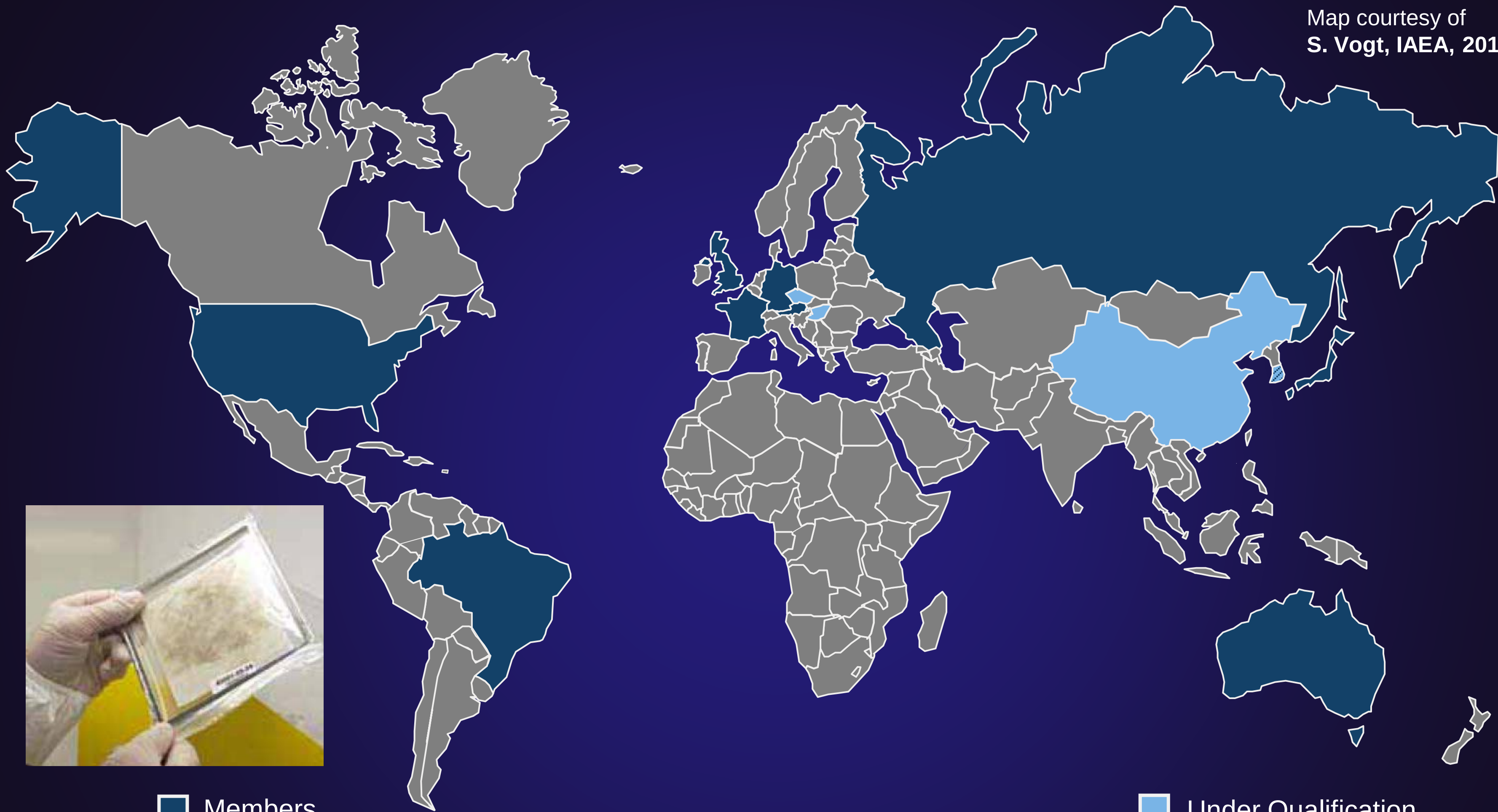
Results: Estimated Source Terms

Sample Number	$^{240}Pu/^{239}Pu$	Std Dev	Approx. % Pu from Global Fallout	Approx. % Pu from NTS Fallout
Soil 1	0.1423	0.0022	75%	25%
Soil 2	0.1467	0.0014	78%	22%
Soil 3	0.1461	0.0016	77%	23%
Soil 4	0.1427	0.0022	75%	25%
Soil 5	0.0871	0.0016	28%	72%

Conclusion: All plutonium in these 5 soil samples is entirely consistent with a mix of global and localized fallout from nuclear weapons testing, and does not indicate any contamination from LANL operations.

IAEA Environmental Sample Analysis Laboratories

Map courtesy of
S. Vogt, IAEA, 2014



Members (legend: dark blue)

IAEA – **SAL**
Australia – **ANSTO, UWA**
Brazil – **IRD**
European Commission – **ITU**
France – **CEA**
Japan – **JAEA**
Republic of Korea – **KAERI**
Russian Federation – **KRI, LMA**
United Kingdom – **AWE**
United States of America – **AFTAC, DOE**

Under Qualification (legend: light blue)

China – **CIAE**
Czech Republic – **UJV Rez**
Hungary – **MTA**
Republic of Korea – **KAERI**

Information Sharing

- **Nuclear forensics can benefit from information sharing**
  - Facilitated by bilateral or multilateral agreements
  - Exchange of experiences or lessons learned
  - Development and use of national libraries
  - Laboratory analysis and data evaluation procedures
  - Training or exercise opportunities

  - *Identify connections between trafficking cases based on material characteristics*
  - *Use the national nuclear forensics library query process to identify possible foreign origins and help address nuclear security issues*

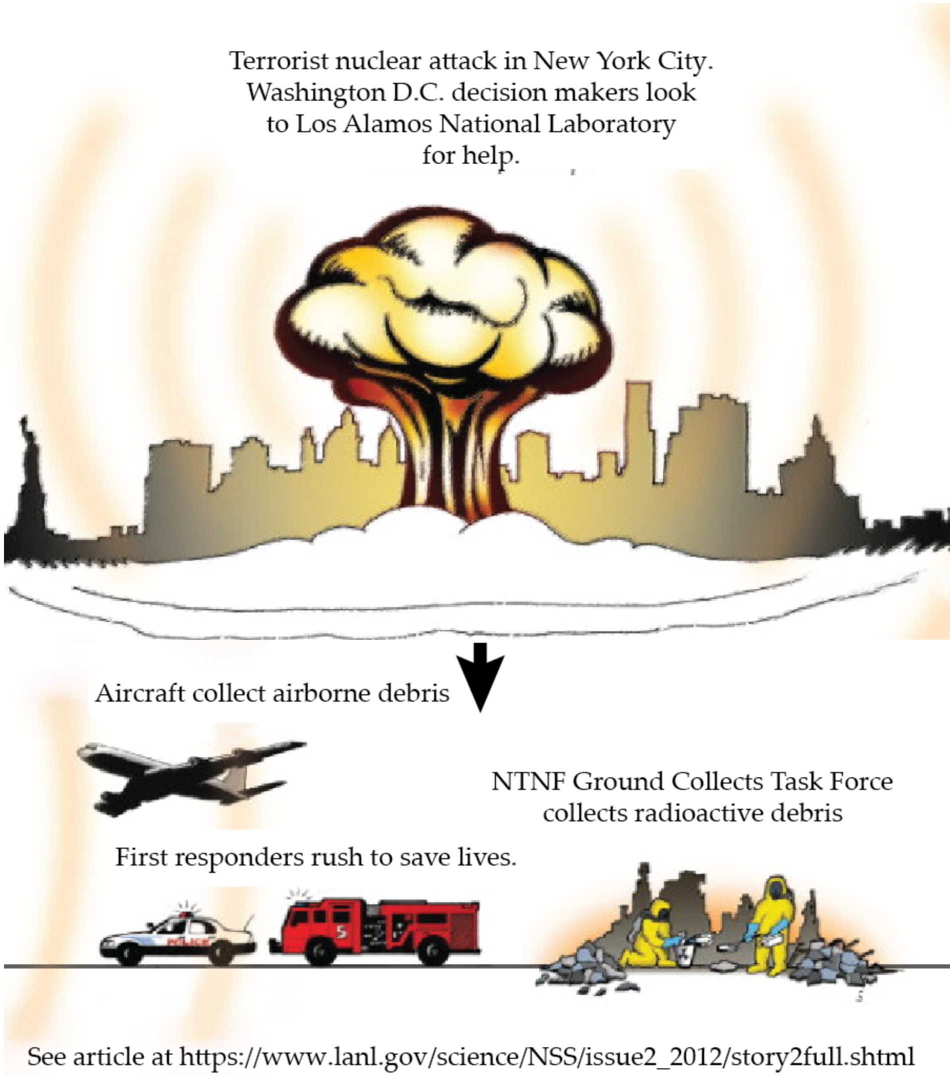
Summary

Nuclear forensics assists in responding to any event where nuclear material is found outside of regulatory control

- Evidence useful for prosecution of traffickers
- Investigative leads for connecting cases, individuals to material, an identifying nuclear security issues

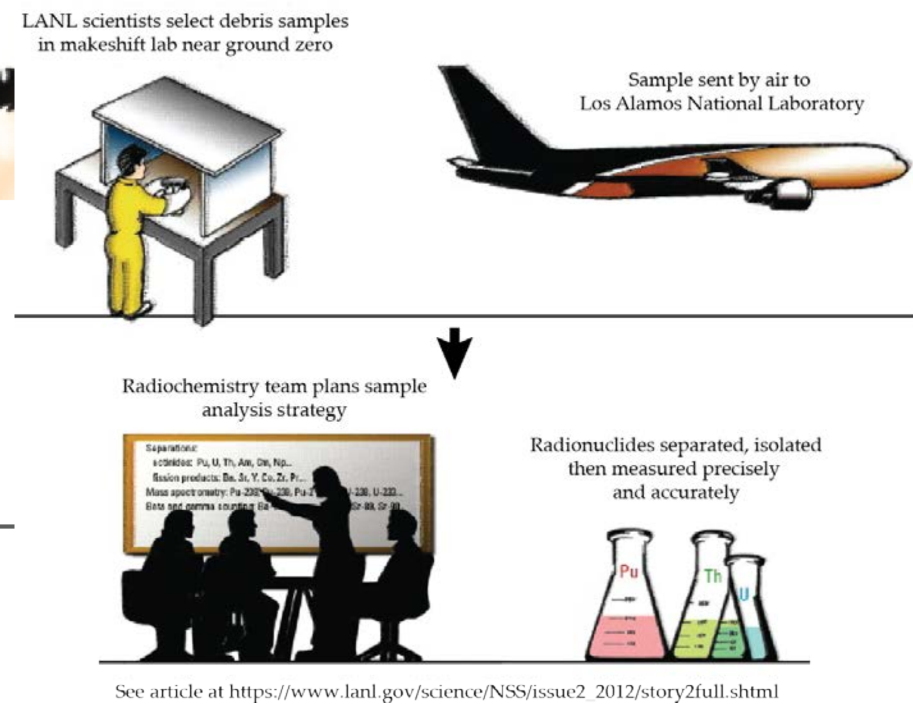
- **Good preparation is essential**
  - National response plan
  - National Nuclear Forensics Library
  - Solid relationships between law enforcement and nuclear forensics laboratories
- **Development of a nuclear forensics program is also a deterrent to smugglers**

Post-detonation Nuclear Forensics



Nuclear terrorism "is the single biggest threat to U.S. security" in " the short term, medium term, and long term"

-- U.S. President Barack Obama

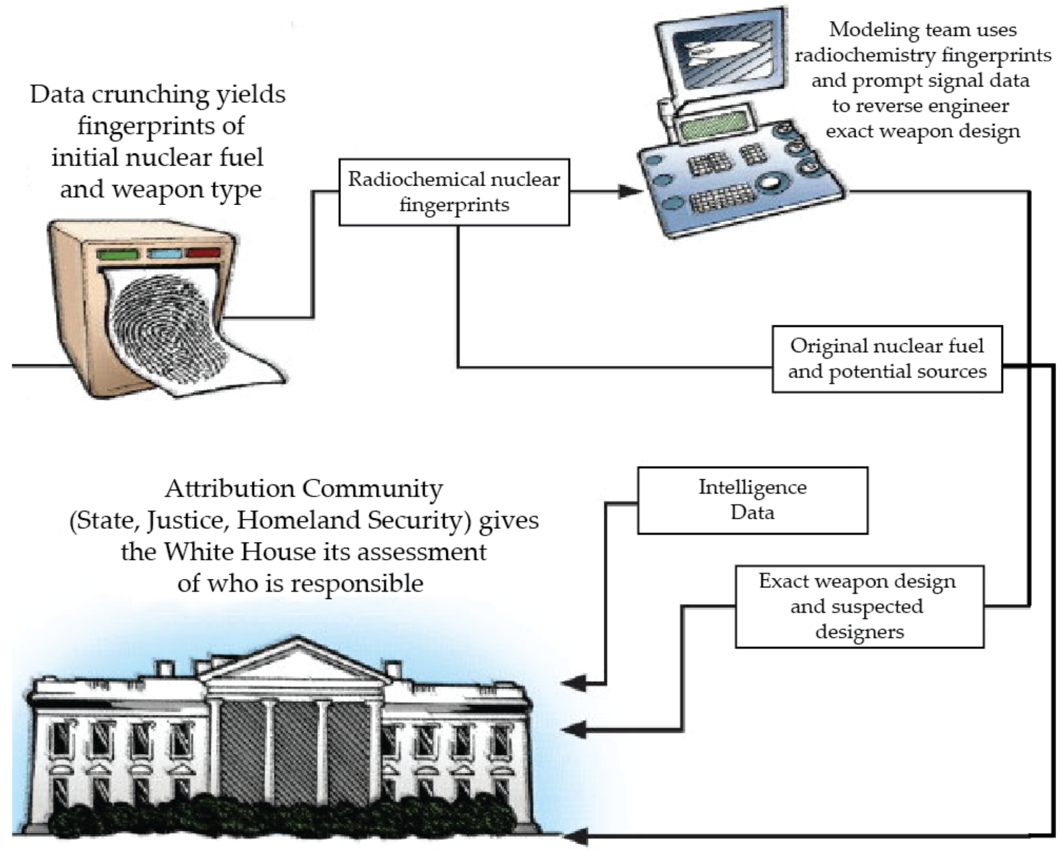


Post-detonation Nuclear Forensics

Design?

Fuel Source?

Inferring the make up of the original fuel (of the bomb) is "unbaking the cake" – **Charles McMillan, Director of Los Alamos National Laboratory**



See article at https://www.lanl.gov/science/NSS/issue2_2012/story2full.shtml